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Collisions between Ultracold Molecules and Atoms in a Magnetic Trap

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We prepare mixtures of ultracold CaF molecules and Rb atoms in a magnetic trap and study their inelastic collisions. When the atoms are prepared in the spin-stretched state and the molecules in the spin-stretched component of the first rotationally excited state, they collide inelastically with a rate coefficient $k_2 = (6.6 \pm 1.5) \times 10^{-11} \text{ cm}^3/\text{s}$ at temperatures near $100 \mu\text{K}$. We attribute this to rotation-changing collisions. When the molecules are in the ground rotational state we see no inelastic loss and set an upper bound on the spin-relaxation rate coefficient of $k_2 < 5.8 \times 10^{-12} \text{ cm}^3/\text{s}$ with 95% confidence. We compare these measurements to the results of a single-channel loss model based on quantum defect theory. The comparison suggests a short-range loss parameter close to unity for rotationally excited molecules, but below 0.04 for molecules in the rotational ground state.

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The formation and control of ultracold molecules is advancing rapidly, motivated by a broad range of exciting applications [1] including tests of fundamental physics, the exploration of many-body quantum physics, quantum information processing, and the study and control of chemical reactions at the quantum level. Collisions are crucial to this field, just as they have been for the field of ultracold atoms. They are a rich source of information about the interactions and reactions of atoms and molecules in a fully quantum-mechanical regime where the internal states of the reactants and the partial waves describing their relative motion are all resolved. Their control is important for evading losses and controlling reactivity [2], and they can be harnessed for sympathetic or evaporative cooling [3,4]. The former is especially important for laser-cooled molecules; having already produced molecules at a few μK [5–7], direct laser cooling is unlikely to lower the temperature much further, and at present the densities are insufficient for evaporative cooling. Instead, sympathetic cooling with evaporatively cooled atoms [8] is a promising way to increase the phase-space density and bridge the gap to quantum degeneracy.

Several methods have been used to study collisions in the temperature range between 10 mK and 1 K. In this cold regime, crossed and merged beams have been used to study quantum state resolved collisions [9–13] and collisions have been investigated in electric and magnetic traps loaded by buffer-gas cooling, Stark deceleration and Zeeman deceleration [14–18]. However, these methods are not suitable for studying collisions at μK temperatures. Molecules in this ultracold regime have been produced by atom association [19], optoelectrical cooling [20], and

direct laser cooling [21,22]. For molecules produced by atom association, molecule-molecule collisions in optical traps have been an important topic of study. These collisions lead to rapid trap loss either due to chemical reactions [2] or, when reactions are energetically forbidden, due to the formation of long-lived complexes that are subsequently excited by the trapping laser [23,24]. The loss rate coefficients are found to be close to those predicted by a single-channel model with universal loss, in which molecules are lost with unit probability once they reach short range [25,26]. Recently, reactive losses of KRb molecules have been suppressed by using an electric field and confining the molecules to two dimensions, and this has led to the formation of a stable quantum degenerate gas of these molecules [4,27,28]. Collisions have also been studied between ultracold CaF molecules in tweezer traps [29,30]. Rapid inelastic losses were observed in these experiments too, both for ground-state molecules and those in excited hyperfine and rotational states. Again, the loss rate coefficient was not far from the one predicted by the universal loss model.

Ultracold atom-molecule collisions have been investigated extensively by theory [31–36], but there are very few experimental studies. Recently, elastic collisions between optically trapped Na atoms and NaLi molecules produced by atom association were observed and used for sympathetic cooling of the molecules [3]. Similarly, elastic collisions of KRb molecules with K atoms have been shown to maintain thermal equilibrium in the formation of a quantum-degenerate Fermi gas [37]. Mixtures of laser-cooled molecules and atoms present exciting new opportunities to study and exploit ultracold collisions. Here, we

produce the first such mixture and use it to study inelastic atom-molecule processes in the μK regime. We produce laser-cooled CaF molecules and Rb atoms, load them into a magnetic trap, and measure the collision-induced loss rate from the trap. When the molecules are in a rotationally excited state, the presence of Rb increases their loss rate, which we attribute to fast rotation-changing collisions. The loss rate coefficient is close to the value predicted by a single-channel universal loss model and is not suppressed when the atoms and molecules are in spin-stretched states. By contrast, when the molecules are in the ground rotational state, no collision-induced losses are observed. We use this observation to set an upper limit to the spin-relaxation rate coefficient. Our development of atom-molecule mixtures, and our study of the inelastic processes within these mixtures, are important steps towards sympathetic cooling.

The starting point of the experiments is a dual-species magneto-optical trap (MOT) of CaF molecules and ^{87}Rb atoms. Each experiment begins by accumulating Rb atoms from a 2D MOT into the dual-species 3D MOT at a rate of about 2×10^9 atoms/s. Once the desired number of atoms has been loaded, the 2D MOT is turned off and the CaF MOT is loaded using the methods described previously. In brief, a pulse of CaF molecules with a mean speed of about 160 m/s is produced by a cryogenic buffer-gas source [38], decelerated to low speed by frequency-chirped laser slowing [39], then captured into a dc MOT [40,41]. To lower the temperature of the molecules, we ramp down the intensity of the main CaF cooling laser to 20% of its initial value over 4 ms, then hold it at this value for 10 ms. An image of the CaF MOT is acquired during this 10 ms period, which we use to determine the initial number of molecules, $N_{\text{CaF}}^{\text{MOT}}$. Next, the quadrupole magnetic field is turned off and both species are cooled simultaneously in two independent optical molasses for 10 ms. For CaF, we follow the molasses procedure described in Ref. [40]. For Rb we linearly ramp the detuning and intensity of the cooling light to -58 MHz and 0.36 mW cm^{-2} over the 10 ms period. The molecules cool to $100 \mu\text{K}$, and the atoms to $40 \mu\text{K}$.

Next, we prepare the molecules in a single, selected quantum state $|N, F, M_F\rangle$ by optical pumping and microwave transfer [42] in an applied magnetic field of 230 mG along z . Here, N is the rotational quantum number and F and M_F are the quantum numbers for the total angular momentum and its projection onto z . At the same time, the atoms are optically pumped into the state $|F = 2, M_F = 2\rangle$. Then, all the laser light is blocked using mechanical shutters and the magnetic quadrupole trap is turned on for a time t at an axial field gradient of 30 G/cm. The trap depth is 1.5 mK. We measure the final number of molecules, $N_{\text{CaF}}(t)$, by recapturing them in the MOT and imaging their fluorescence for 10 ms. Molecules prepared in $N = 0$ are transferred back to $N = 1$ using a microwave pulse prior to recapture in the MOT. We measure the final

number of atoms, $N_{\text{Rb}}(t)$, by taking an absorption image of the cloud shortly after releasing it from the magnetic trap. The collisional loss rate is determined by measuring the fraction of molecules remaining, $r_{\text{CaF}}(t) = N_{\text{CaF}}(t)/N_{\text{CaF}}^{\text{MOT}}$, as a function of t , both with and without atoms in the trap. Division by $N_{\text{CaF}}^{\text{MOT}}$ makes the measurements immune to shot-to-shot fluctuations in the number of molecules in the MOT. Our simulations of the trap loading show that the sizes and positions of the clouds reach a steady state within 300 ms, which is the minimum value of t we use.

The density of the atomic sample exceeds that of the molecules by six orders of magnitude, so we can ignore molecule-molecule collisions and loss of atoms due to collisions with molecules. In this case, the loss of molecules from the trap is described by

$$\dot{r}_{\text{CaF}} = (-\Gamma_{\text{CaF}}^0 - \Gamma_{\text{Rb-CaF}} e^{-\Gamma_{\text{Rb}} t}) r_{\text{CaF}}, \quad (1)$$

whose solution is

$$r_{\text{CaF}}(t) = r_{\text{CaF}}(0) \exp \left(-\Gamma_{\text{CaF}}^0 t - \frac{\Gamma_{\text{Rb-CaF}}}{\Gamma_{\text{Rb}}} (1 - e^{-\Gamma_{\text{Rb}} t}) \right). \quad (2)$$

Here Γ_{CaF}^0 is the loss rate of CaF in the absence of Rb, Γ_{Rb} is the loss rate of Rb, and $\Gamma_{\text{Rb-CaF}}$ is the collision-induced loss rate of CaF at $t = 0$. This is

$$\Gamma_{\text{Rb-CaF}} = k_2 N_{\text{Rb}}(0) \int f_{\text{Rb}}(\vec{r}) f_{\text{CaF}}(\vec{r}) d^3 \vec{r} = k_2 \zeta, \quad (3)$$

where k_2 is the inelastic rate coefficient and $f_s(\vec{r})$ is the density distribution of species s normalized such that $\int f_s(\vec{r}) d^3 \vec{r} = 1$. The equation defines ζ , an effective Rb density that accounts for the overlap between the two distributions.

Figure 1 is an example of one measurement showing the fraction of molecules remaining in the trap as a function of t , for molecules prepared in the state $|1, 2, 2\rangle$. In the absence of Rb (filled points), the data fit well to a single-exponential decay with a loss rate of $\Gamma_{\text{CaF}}^0 = 0.35(1) \text{ s}^{-1}$. This is the natural loss rate due to collisions with residual background gas and vibrational excitation by blackbody radiation [42]. The Rb data, $N_{\text{Rb}}(t)$, also fit well to a single exponential with $\Gamma_{\text{Rb}} = 0.313(5) \text{ s}^{-1}$. These single-species loss rates vary negligibly throughout the experiments. The open points show the loss of molecules in the presence of $N_{\text{Rb}}(0) = 1.50(15) \times 10^9$ atoms with a peak number density of $5(1) \times 10^{10} \text{ cm}^{-3}$. We fit these data to Eq. (2) with $r_{\text{CaF}}(0)$ and $\Gamma_{\text{Rb-CaF}}$ as free parameters. We account for the uncertainties in Γ_{CaF}^0 and Γ_{Rb} using the method described in the Supplemental Material [43], which yields the distribution of $\Gamma_{\text{Rb-CaF}}$ values shown in the inset of the figure. The mean and standard deviation of this distribution give our best estimate for this dataset, $\Gamma_{\text{Rb-CaF}} = 0.368(44) \text{ s}^{-1}$.

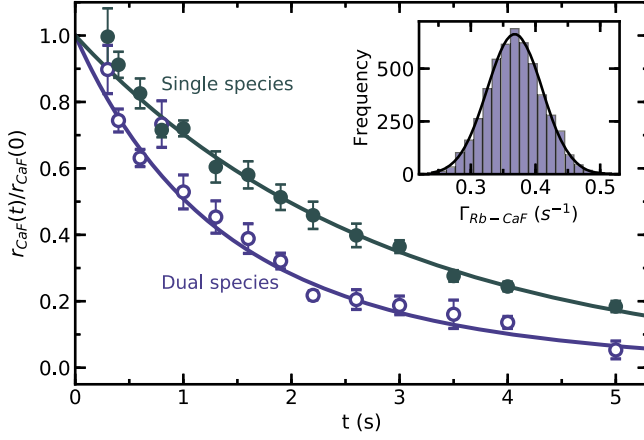


FIG. 1. Fraction of molecules, $r_{\text{CaF}}(t)/r_{\text{CaF}}(0)$, remaining in the magnetic trap after a hold time t , with Rb (open points) and without Rb (filled points). The molecules are prepared in the $|1, 2, 2\rangle$ state. Each point is the average and standard deviation of six measurements. For the single species data the line is a fit to an exponential decay model. For the two-species data the line is a fit to Eq. (2). Inset: histogram of $\Gamma_{\text{Rb-CaF}}$ estimates using the method described in [43], together with a fit to a normal distribution.

To determine ζ , we measure the density distributions of both species in the magnetic trap by turning off the magnetic field gradient and then imaging their distributions. For Rb we use absorption imaging as described in [43]. For CaF, we turn on the MOT light for 1 ms, with the field gradient still off, and image the fluorescence. Expansion of the cloud during this short period is negligible. We fit the radial distributions to Gaussians and the axial distributions to the equilibrium distribution for a magnetic quadrupole trap plus gravity [43]. We use these fits to calculate the overlap integral in Eq. (3). Our choice of axial distribution automatically accounts for the differential gravitational sag between the two clouds. The temperatures in the magnetic trap are measured using the standard ballistic expansion technique. We find that, when the CaF is loaded into the trap, its $1/e^2$ radius expands from 1.6(1) to 2.30(7) mm and its geometric-mean temperature rises to 175(37) μK . This is because the initial position of the CaF cloud is displaced from the trap center. The Rb cloud is well centered because of the greater flexibility in controlling the intensity balance of the Rb MOT light. Its geometric-mean temperature in the magnetic trap is 71(1) μK .

We repeat the measurements of $\Gamma_{\text{Rb-CaF}}$ and ζ for various values of $N_{\text{Rb}}(0)$. The Rb density distribution, f_{Rb} , depends on the number of atoms, so we measure it in every case. Figure 2 shows how $\Gamma_{\text{Rb-CaF}}$ varies with ζ , together with the measured radial cloud distributions for the smallest and largest atom clouds used. The gradient of the straight-line fit gives the value of k_2 . We account for the uncertainties in the measured number of atoms and cloud sizes using the method described in [43]. The result is

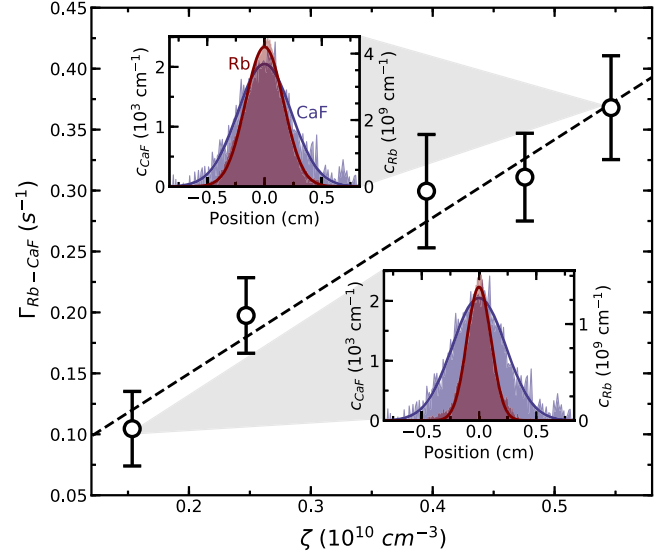


FIG. 2. Points: collisional loss rate, $\Gamma_{\text{Rb-CaF}}$, as a function of effective density, ζ , for molecules in the $|1, 2, 2\rangle$ state. Line: straight line fit to the data. Insets: radial density distributions, $c_s(x) = \int N_s f_s(x, y, z) dy dz$, of the atoms and molecules for small and large ζ , with Gaussian fits.

$k_2 = (6.6 \pm 1.5) \times 10^{-11} \text{ cm}^3/\text{s}$ for the state $|1, 2, 2\rangle$. As discussed later, we attribute this loss to collisions with Rb that change the rotational state of the molecule from $N = 1$ to $N = 0$. This results in a loss of molecules for two reasons: (i) we detect molecules only in $N = 1$; (ii) the collision releases 1 K of energy, which is far greater than the trap depth.

For the measurement described above, both the atoms and molecules are in spin-stretched states, an arrangement that often suppresses inelastic collisions [3,14]. To test whether that is the case here, we repeat the measurement using molecules in the $|1, 1^+, 1\rangle$ state [51], which is not spin stretched. We obtain $k_2 = (5.7 \pm 1.8) \times 10^{-11} \text{ cm}^3/\text{s}$, showing that these rotation-changing collisions have no strong dependence on the choice of hyperfine or Zeeman level.

Next, we make the same measurement for molecules prepared in the ground rotational state $|0, 1, 1\rangle$. Here, the only open loss channel is spin relaxation. Figure 3 compares the loss rates for molecules in $N = 0$ and $N = 1$, in the presence of $N_{\text{Rb}}(0) = 1.2 \times 10^9$ atoms, dramatically illustrating how the choice of rotational state influences these ultracold inelastic collisions. For these data, the collisional loss rate is $0.30(5) \text{ s}^{-1}$ for molecules in $|1, 2, 2\rangle$ and $0.013(36) \text{ s}^{-1}$ for molecules in $|0, 1, 1\rangle$. Averaging several similar measurements we obtain $k_2 = -0.05 \pm 2.9 \times 10^{-12} \text{ cm}^3/\text{s}$. This result is consistent with zero and yields the upper bound $k_2 < 5.8 \times 10^{-12} \text{ cm}^3/\text{s}$ with 95% confidence. This is an order of magnitude smaller than the rotation-changing rate coefficient for molecules in the first excited rotational state.

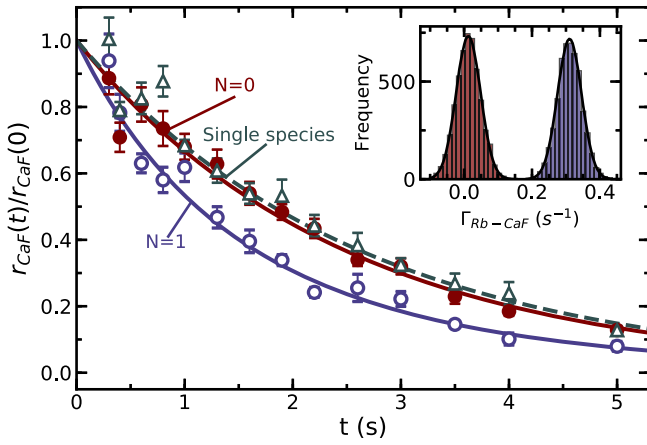


FIG. 3. Loss of molecules from the magnetic trap in the presence of 1.2×10^9 atoms, for molecules in the $|1, 2, 2\rangle$ state (open points) and the $|0, 1, 1\rangle$ state (filled points). Solid lines: fits to Eq. (2). Also shown is the loss of molecules in $|0, 1, 1\rangle$ in the absence of Rb (open triangles) together with a single exponential fit (dashed line). Inset: histogram of $\Gamma_{\text{Rb-CaF}}$ estimates, together with fits to normal distributions.

To interpret the experimental loss rates, we compare to calculations with a single-channel nonuniversal model based on quantum defect theory [26,43]. This models the long-range interaction potential by its asymptotic form $-C_6 R^{-6}$ and takes full account of temperature dependence, which is important under the conditions of the experiment. It has been used successfully to model losses in collisions of ultracold RbCs molecules [52]. The model characterizes the complicated short-range part of the interaction in terms of two parameters, y and δ^s . The loss parameter y runs from 0 (no loss) to 1 (complete loss at short range, corresponding to a universal model). It accounts for all sources of collisional loss, including rotational inelasticity and spin-changing collisions. δ^s is the phase shift between the incoming wave and the reflected wave at short range, and is related to the scattering length.

Figure 4 shows a contour plot of the rate coefficient obtained from the single-channel model as a function of y and δ^s . The experimental value and range of uncertainty for $|1, 2, 2\rangle$ and the upper limit for $|0, 1, 1\rangle$ are indicated as lines. The calculated loss rates show significant peaks around $\delta^s = 3\pi/8$ and $5\pi/8$ and a weaker peak around $7\pi/8$; these are associated with p -, d -, and f -wave shape resonances, respectively. The corresponding s -wave peak around $\delta^s = \pi/8$, which is very prominent at lower temperatures [26,52], is weakened at these temperatures due to increased transmission past the long-range potential and by thermal averaging.

The measured loss rate for molecules in $N = 1$ is close to the universal rate, which is $8.2 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$ at this temperature. The most likely ranges of parameters have $y > 0.4$, but the measurements do not rule out significantly lower y if there is resonant enhancement. Since the spin

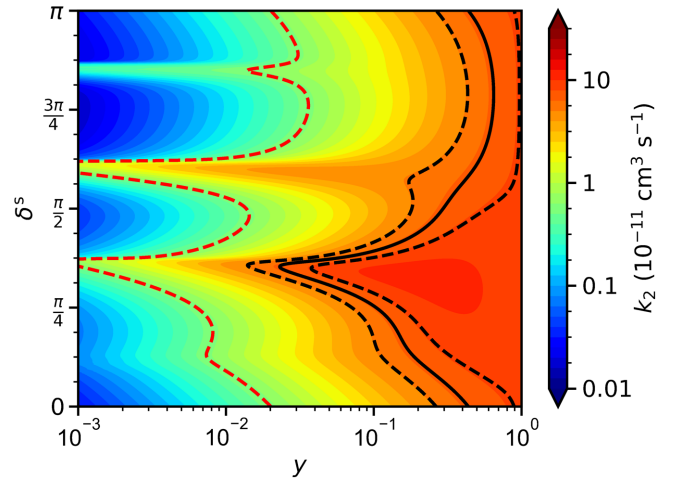


FIG. 4. Thermally averaged loss rate coefficient k_2 from the single-channel model as a function of loss parameter y and short-range phase shift δ^s . The mean and standard error of the experimentally measured k_2 for molecules in $|1, 2, 2\rangle$ are shown as black lines. The 95% upper bound for k_2 for molecules in $|0, 1, 1\rangle$ is shown as a dashed red line.

state is found to have little influence on the loss rate and complex-mediated loss is unlikely to be important for this system [43], we conclude that this loss is dominated by rotation-changing collisions. For the rotational ground state, the measured loss rate is consistent with loss parameters $y < 0.04$. Overall, the results are consistent with the expectation that rotational relaxation is very fast, but that spin relaxation is slow.

Full coupled-channel scattering calculations including nuclear spin [35] are not feasible for Rb + CaF, because the very deep potential well would require an impractically large basis set for convergence. Even without nuclear spin, they are very challenging. Morita *et al.* have carried out such calculations on a single interaction potential for the related system Rb + SrF. However, the particular interaction potential they used produces a p -wave resonance very close to threshold; it would correspond to $\delta^s \approx 3\pi/8$ in our model. As shown in Fig. 4, such a potential is likely to produce atypically large loss rates. At a collision energy of 10^{-4} cm^{-1} and a magnetic field of 1 G, their calculation gave a spin-relaxation cross section of $\sim 200 \text{ \AA}^2$; without thermal averaging, this corresponds to $k_2 \approx 5 \times 10^{-13} \text{ cm}^3 \text{ s}^{-1}$, which is well within our experimental upper bound.

In summary, we have produced the first mixtures of laser-cooled atoms and molecules and have studied their inelastic collisions in the ultracold regime. We have compared the results to a short-range loss model and find that rotational relaxation proceeds rapidly, close to the universal rate and independent of hyperfine and magnetic sublevel, whereas spin relaxation is at least 10 times slower. The latter observation is encouraging for the prospects of sympathetic cooling, where spin relaxation processes are expected to be a limiting factor.

Our mixture can be used to investigate collisions in other settings. For example, studies in the dual-species MOT can shed light on inelastic collisions with excited-state atoms and light-assisted processes [53]. By capturing atoms and molecules from the mixture into tweezer traps, the collisions can be studied at the single-particle level [30]. It will be interesting to investigate the influence of applied electric and magnetic fields on these collisional processes [15,35]. The mixture could also be used to form ultracold triatomic molecules by photoassociation or magnetoassociation. We are currently working to trap more molecules, trap atoms at a higher density, improve the overlap of the clouds, and incorporate an optical dipole trap. With these improvements, we aim to study the elastic collisions required for sympathetic cooling of molecules by evaporatively cooled atoms [8], which is a promising way to increase the phase-space density towards quantum degeneracy.

Underlying data may be accessed from Zenodo [54] and used under the Creative Commons CCZero license.

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